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Positronium collisions with atoms and molecules

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Abstract. We review recent theoretical efforts to explain observed similarities between electron-atom and positronium(Ps)-atom scattering which also extends to molecular targets. In the range of the projectile velocities above the threshold for Ps ionization (break-up) this similarity can be explained in terms of quasi-free electron scattering and impulse approximation. However, for lower Ps velocities more sophisticated methods should be developed. Our calculations of Ps scattering by heavy noble-gas atoms agree well with experiments at Ps velocities above the Ps ionization threshold. However, in contrast to electron scattering cross sections, at lower velocities they exhibit maxima whereas the experimental cross sections tend to decrease toward lower velocities indicating the same similarity with electron scattering cross section observed above the threshold. Our preliminary results for Ps-N₂ scattering confirm experimental observation of a resonance similar to the Π_g resonance in electron-N₂ scattering.

1. Introduction

Recent measurements [1, 2, 3, 4, 5] of positronium (Ps) collision cross sections for a number of atoms and small molecules show interesting similarities between the Ps scattering and the electron scattering in the intermediate energy range. When plotted as a function of the projectile velocity, the electron and Ps cross sections are very close and even show similar resonance-like features. Several theoretical efforts [6, 7, 8, 9] were directed to explain this intriguing phenomenon.

To understand the physics of these similarities, we first have to identify the processes which contribute to the measured total cross sections for collisions of Ps with an atomic or molecular target A . These are elastic scattering

$$\text{Ps}(1s) + A \rightarrow \text{Ps}(1s) + A,$$

Ps excitation

$$\text{Ps}(1s) + A \rightarrow \text{Ps}(nl) + A,$$

and Ps ionization (or break-up):

$$\text{Ps}(1s) + A \rightarrow e^- + e^+ + A.$$



Since the atomic and molecular targets of interest typically contain tightly bound electrons, excitation and ionization of the target can be neglected. In addition it was shown [6, 10, 11] that the cross sections for excitation of Ps are typically small compared to cross sections for ionization. Therefore the main processes contributing to the total cross section are elastic scattering and ionization of Ps.

It is well known [12, 13] that except for very low projectile velocities the electron scattering cross sections are much larger than the positron scattering cross sections for the same target. This is due to two factors: First, the static and polarization potentials for electron-atom interaction are both attractive, whereas for the positron-atom interaction the former is repulsive and the latter is attractive, and they partly cancel each other. Second, there is an additional exchange interaction in the case of electron-atom scattering whereas it is absent in positron-atom scattering. This allows one to suggest that Ps-atom scattering is dominated by the electron-atom interaction and can be described in terms of electron-atom scattering.

There is a close analogy between this situation and that which occurs in Rydberg-atom-ground-state-atom collisions. According to the quasi-free electron model, suggested initially by Fermi [14] and developed further by Matsuzawa [15], Rydberg-atom-ground-state atom scattering amplitudes can be described in terms of the free-electron scattering amplitude. Although the effect of scattering of the ion core of the Rydberg atom might be significant as well, these two processes, i.e. electron scattering and ion-core scattering, can be treated independently. There is a particularly simple relation between the corresponding cross sections at thermal collision velocities [15]. In this case the velocity of the Rydberg electron in the laboratory frame is essentially equal to the orbital velocity in the Rydberg atom, and the cross section for the Rydberg-atom-ground-state atom collision can be expressed as the electron scattering cross section averaged over the Rydberg electron velocity distribution.

However, such a simple approach is not valid for Ps-atom collisions. First, the electron in the ground state of a Ps atom, although less strongly bound than in the hydrogen atom, can be hardly treated as a quasi-free particle. Second, a typical Ps collision velocity is comparable to the electron velocity in Ps, therefore a simple average over the velocity distribution is not possible. Therefore for a theoretical description of Ps-atom and Ps-molecule collisions more advanced approaches are needed.

The rest of this article contains a summary of the theoretical approaches which we have used recently for the description of Ps-atom and Ps-molecule collisions with some specific examples illustrating the similarity between electron and Ps scattering. For simplicity in what follows we will often refer to Ps-atom scattering, but many statements will be applicable to Ps-molecule scattering as well.

2. Impulse and binary-encounter approximations

A firmer theoretical ground for the Fermi picture is provided by the Faddeev-equation method [16] allowing formulation of an equation for the scattering amplitude for a composite projectile in terms of the scattering amplitudes for its constituents. The first approximation for the Faddeev amplitude leads to the impulse approximation. The impulse approximation serves as a solid theoretical foundation for the Fermi model and does not require the projectile velocity to be small compared to the orbital velocities of the constituent particles. However, a weak binding is still required. We can assume that this requirement is not so strict because of the large difference between the amplitudes for electron-atom and positron-atom scattering. This assumption allows us to express the Ps-atom cross section in terms of the electron-atom scattering amplitudes. It is further assumed that the Ps energy is well above the Ps ionization (break-up) threshold. These two assumptions allowed us to prove [6] that the Ps-atom cross section equals the electron scattering cross section for equal incident velocities. Moreover, calculations of cross sections for Ps collisions with noble-gas atoms employing the impulse approximation show good agreement

with experiment above the ionization threshold which corresponds to the Ps velocity 0.5 a.u. [6].

However, both assumptions involved in these calculations become invalid for projectile velocities below the Ps ionization threshold. Indeed, for lower energies the Ps-atom cross section increases sharply toward lower energies, in contrast to experimental data.

Another problem with the impulse approximation is that it employs off-shell scattering amplitude. To avoid this complication, an on-shell reduction is typically used [17]. However, this reduction is not unambiguous. A more physical, albeit a more simplified, approach is the binary-encounter approximation [18, 19] where it is assumed that the electron and positron interact with the target independently, but incoherently. The cross section for Ps ionization can be obtained by employing on-shell scattering amplitudes for electron-atom and positron-atom elastic scattering. This approach was successfully used for calculations of ionization of Ps in collisions with noble-gas atoms [9] and the hydrogen molecule [8], and the results are close to those of the more sophisticated impulse approximation [17].

3. Pseudopotential method

Although the impulse approximation fails at low energies, the main idea that Ps-atom scattering can be described in terms of electron-atom and positron-atom scattering phase shifts can also be used in the pseudopotential method [7]. It involves constructing model potentials that reproduce these phase shifts, and then adding them to describe the Ps-atom interaction. While this procedure is straightforward for positron scattering, the situation with electrons is more complicated. Due to the Pauli exclusion principle, the effective potential for the electron depends on its orbital angular momentum l , i.e., it becomes a pseudopotential [20]. Formally this means that the effective electron-atom potential is a nonlocal operator with the kernel

$$V_e(\mathbf{r}, \mathbf{r}') = \frac{1}{r^2} \delta(r - r') \sum_{lm} V_l(r) Y_{lm}^*(\hat{\mathbf{r}}) Y_{lm}(\hat{\mathbf{r}}'), \quad (1)$$

where $Y_{lm}(\hat{\mathbf{r}})$ is a spherical harmonic. When such a pseudopotential is averaged over the electron density distribution in the Ps atom, it becomes a nonlocal operator. The phase shifts necessary to construct the pseudopotential should be calculated in the static-exchange (for electrons) or static (for positrons) approximation. This is because the exact phase shifts incorporate the long-range polarization interaction between the electron or positron and the target decaying as $1/r^4$ at large distances r . However, the long-range interaction between the Ps and a neutral target is due to the van der Waals force which decreases as $1/R^6$ at large Ps-atom distances R . Therefore it should be added to the Ps-atom pseudopotential. We do this in the form of the potential

$$V_W(R) = -\frac{C_6}{R^6} \left\{ 1 - \exp[-(R/R_c)^8] \right\}, \quad (2)$$

where R_c is an adjustable short-range cut-off radius, and C_6 is the van der Waals constant¹. Parameter R_c depends on the short-range correlations whose contribution to Ps-atom scattering is difficult to calculate *ab initio*. We choose it by requiring that at higher energies the results of the pseudopotential calculations merge with those of the impulse approximation. Alternatively R_c can be treated simply as an empirical parameter.

4. Scattering by noble-gas atoms: Ramsauer-Townsend minimum?

Calculations based on the pseudopotential method [7, 8, 9] show good agreement with experiments [1, 3] above the Ps ionization threshold and confirm the similarity between Ps-atom

¹ See Ref. [21] for the accurate values of C_6 for Ps-atom pairs for noble gases and a number of other atoms.

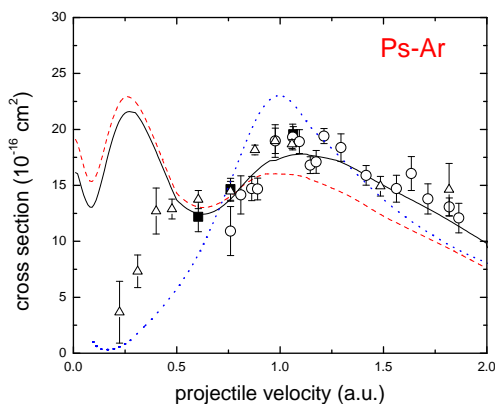


Figure 1. Ps-Ar total scattering cross sections. Solid (black) and dashed (red) curves are results of the pseudopotential calculations [7] with $R_c = 2.5$ and 3.0 a.u. respectively. Experiment: \circ [23]; \blacksquare [1]; \triangle [4]. Dotted blue line, e -Ar elastic scattering cross section compiled from the calculations [24] and measurements [25].

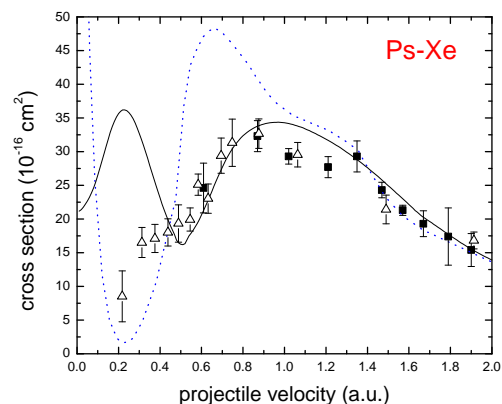


Figure 2. Cross sections for Ps-Xe collisions. Theory [9]: solid (black) line; Experiment: \blacksquare [1]; \triangle [4]. Dotted blue line, e -Xe elastic scattering cross section compiled from the calculations [26] and measurements [27].

and electron-atom scattering. They also show that at low Ps velocities the Ps-atom interaction is dominated by an effective repulsion due to the Pauli exclusion principle. The long-range van der Waals interaction is substantially weaker, and this leads to positive scattering lengths for Ps collisions with Ar, Kr and Xe. These results are confirmed by Ps-atom scattering calculations [22] in which the internal electron-positron dynamics inside the Ps atom is fully accounted for and which use the same van der Waals potential. In contrast, the electron interaction with heavy noble-gas atoms is dominated by the long-range polarization attraction, and this leads to negative scattering lengths and the appearance of the Ramsauer-Townsend minimum in the scattering cross sections. Therefore our conclusion [7, 9] was that the similarity between electron and Ps scattering by heavy noble-gas atoms disappears at low collision energies. However, this conclusion was not confirmed by recent measurements [4] of Ps-Ar and Ps-Xe scattering cross sections which continue to decrease towards lower energies and indicate the existence of a minimum in these cross sections.

In Figs. 1 and 2 we present comparisons of theoretical results with measured results showing that the theory and experiment demonstrate opposite trends when the Ps velocity decreases below the Ps break-up threshold ($v = 0.5$ a.u.). Moreover, theoretical results show a maximum instead of a minimum which is particularly pronounced in Ps-Xe collisions. This maximum is mostly due to the p -wave contribution and could result from a too fast decrease of the calculated p -wave phase shift. We believe that an improved treatment of the short-range correlations might eliminate this maximum and improve agreement with the experiment. However, this improvement is very unlikely to change the sign of the scattering length and to lead to the Ramsauer-Townsend minimum. This conclusion is confirmed by independent calculations of Mitroy *et al.* [28, 29] and of Swann and Gribakin [22] who also obtained positive scattering lengths for Ar, Kr and Xe, as well as for the lighter atoms, He and Ne.

5. Scattering by molecules and resonances

Although an extension of the pseudopotential method to Ps-molecule scattering is straightforward, it is becoming computationally very challenging, particularly for polyatomics. Two approaches can be explored in this case. First, if the electron-molecule and positron-molecule interaction potentials are close to being spherically symmetric, the pseudopotential method developed for Ps-atom scattering can be applied to molecules without modifications. We used this method to investigate Ps-H₂ collisions [8].

Another approach explored by us generalizes the local exchange potential used for electron-molecule scattering [30] to Ps-molecule scattering. The idea is to treat the target electrons as a free electron gas and use the known expression for the energy of interaction of a projectile electron with the gas electrons. The obtained exchange potential was used to describe Ps-N₂ scattering. The N₂ molecule is interesting because a recent experiment [5] shows that a well-known Π_g resonance observed in e -N₂ scattering is also present in Ps-N₂ scattering.

Our calculations are based on the free-electron-gas exchange potential with added repulsive cores in s - and p -wave components of the scattering wavefunction to mock orthogonality of the projectile electron wave function to the occupied σ_g , σ_u and π_u target orbitals.

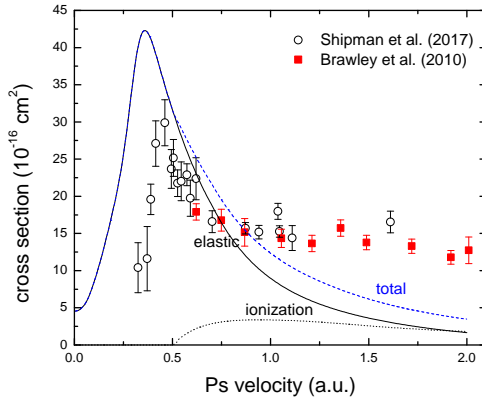


Figure 3. Ps-N₂ scattering cross sections, comparison of the theory with experimental data [1, 5].

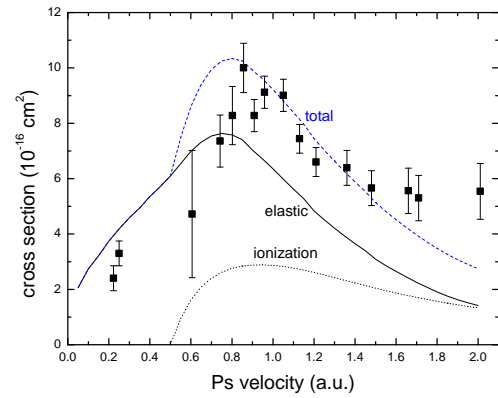


Figure 4. Ps-H₂ scattering cross sections, comparison of the theory [8] with experimental data [31, 32].

Preliminary results are shown in Fig. 3. where we present theoretical elastic, ionization and total cross sections, and compare the latter with the experimental data [1, 5]. The theoretical resonance peak position ($v = 0.365$ a.u.) is slightly shifted towards lower Ps velocities relative to the experimental peak position ($v = 0.46$ a.u.) and the theoretical peak cross section is higher than the experimental one. The latter might be due to the fixed-nuclei approximation used in the present calculations which do not take into account the vibrational dynamics. A significant difference between the theory and experiment is observed at higher velocities where the experimental cross section remains flat and stays close to the e -N₂ cross section (not shown), whereas the theoretical curve is showing a relatively fast decrease with growing v . A similar tendency was observed in Ps-H₂ calculations [8] shown in Fig. 4. The most likely explanation for this disagreement is the absence of short-range correlations in the present version of the theory.

6. Conclusion and outlook

In summary, in the past three years we have exploited several methods to describe similarities between electron-atom and Ps-atom scattering. At a qualitative and semiquantitative levels this similarity can be explained in terms of the impulse approximation which treats electron and positron scattering independently. However, at lower energies the impulse approximation breaks

down, and more sophisticated methods like the pseudopotential approach and free-electron-gas exchange approximation should be developed.

A big challenge to the theory remains the observed [4] behaviour of low-energy Ps-Ar and Ps-Xe scattering showing decrease of cross sections towards lower energies and indicating an existence of the Ramsauer-Townsend minimum similar to that observed in e -Ar and e -Xe scattering. It is hard to explain this behaviour from the theoretical point of view since the long-range van der Waals interaction is too weak to overcome the effective repulsive core due to the electron exchange. On the other hand it is likely that our p -wave contribution which creates a low-energy peak in Ps scattering by heavy noble-gas atoms is too large, and a more accurate account of the exchange and short-range correlations would eliminate this peak (as seen, e.g., in [22]) and could make the theoretical cross sections closer to the experiment.

Our first attempt to describe the resonant Ps-molecule scattering confirms the existence of the observed resonance in Ps-N₂ collisions similar to the Π_g resonance in e^- -N₂ collisions. Future work should include short-range correlations and the nuclear motion.

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